

PROSPECTIVE ASP FIELD PROJECTS

Preliminary Project Descriptors for Initial Consideration

This document consists of brief descriptions of prospective field studies for the Department of Energy Atmospheric Science Program (<http://www.asp.bnl.gov>). These project descriptors are presented here to facilitate discussion of future field studies to be undertaken by the program.

ASP Participation in VOCALS-Regional Experiment (Marine Stratus off Chile/Peru)

Peter Daum, Yangang Liu, BNL

Atmospheric Aerosol Characterization and Calibration Facility

Stephen E. Schwartz (ses@bnl.gov), Yin-Nan Lee; Brookhaven National Laboratory

Carbonaceous Aerosol Evolution (CARE) Campaign

Rahul Zaveri (Rahul.Zaveri@pnl.gov), Chris Doran, Carl Berkowitz, Liz Alexander, Alex Laskin, Jerome Fast, John Hubbe, PNNL; Sasha Madronich, NCAR; Doug Worsnop, Aerodyne; Jose Jimenez, University of Colorado; Joel Thornton, University of Washington; Rainer Volkamer, University of California San Diego

Deep Convective Clouds and Chemistry (DC3)

Steve Ghan, Pacific Northwest National Laboratory (Steve.Ghan@pnl.gov)

Ganges Valley Aerosol Experiment (GVAX)

V. Rao Kotamarthi, Argonne National Laboratory (vrkotamarthi@anl.gov)

Indirect and Semi-Direct Aerosol Campaign (ISDAC)

Steve Ghan, Pacific Northwest National Laboratory, (Steve.Ghan@pnl.gov)

Megacity Aerosol Experiments

Larry Kleinman, BNL (kleinman@bnl.gov)

Rapid deployment capability for characterization of unique events

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The Biospheric Aerosol Production Experiment - BAP

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Time evolution of the optical and chemical properties of smoke from brush and pine forest fires in the relatively dry western US.

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Winter/Summer Aerosol Composition and Evolution Experiment

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ASP Participation in VOCALS-Regional Experiment (Marine Stratus off Chile/Peru)

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BNL proposes that ASP participate in an experiment called VAMOS Ocean-Cloud-Atmospheric-Land Study (VOCALS). VOCALS is an NSF sponsored experiment designed to examine properties and processes occurring in marine stratus clouds off the coast of South America (Chile-Peru). The experiment was originally scheduled for Oct 2007, but it is highly probable that it will be delayed until October 2008. The experiment will be conducted in a climatologically important, but poorly understood region of the globe where extensive areas of marine clouds exist (coverage ~70% during the month of October). West of the coastline, clouds are generally pristine in terms of anthropogenic influence because virtually no upwind (winds generally from the west and southwest) sources of anthropogenic aerosols exist for thousands of miles. Close to coast however, clouds can be influenced by aerosols from urban areas and from industrial sources such as smelters that are located both inland and along the coast. Thus, conditions will prevail such that we can sample the properties of clouds that are truly background, and those that are subjected to varying degrees of anthropogenic influence. This contrasts to the situation that we have encountered in our previous US based cloud studies (NARE and MASE) where clouds that were sampled were nearly always perturbed by substantial quantities of anthropogenic aerosols.

Motivation- VOCALS represents a unique opportunity to examine both first and second indirect aerosol issues (influence of anthropogenic aerosols on drizzle formation) in clouds that range from pristine to those that are heavily influenced by anthropogenic aerosols. The experiment provides a unique opportunity to evaluate our recently developed drizzle parameterizations, and to examine effects of anthropogenic aerosols on cloud microphysics in a region where clouds are thought to be most sensitive to the effects of anthropogenic aerosols.

Strategy- NOAA is tentatively planning to deploy the P-3 in the experiment. We have discussed with Dr. Chuck Brock the possibility of joining that effort, providing aerosol and cloud microphysics instruments and expertise to that effort. Depending on the extent of interest from the ASP community, we could contemplate deploying the G-1 equipped with a full array of instrumentation to measure aerosol and cloud properties.

Collaborators- Base experiment is NSF funded. There will be South American participants. NCAR will fly the C-130; NOAA is proposing to fly the P-3.

Atmospheric Aerosol Characterization and Calibration Facility

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A facility consisting of appropriate inlets for sampling atmospheric aerosols and related gas-phase constituents, and providing a testbed for operation of a suite of instruments for systematic characterizing the microphysical, chemical, optical, and cloud nucleating properties of atmospheric aerosols over an extended period of time.

It would be intended to keep a broad suite of such instruments running for much of the time. Develop broad repertoire of simultaneous measurements of these aerosol properties, including diurnal and seasonal cycles.

Many of these instruments would be those which are deployed in ASP field studies but which see little use otherwise; this facility would greatly enhance the data output of such instruments; also experience in operating these instruments and maintaining calibration would assure maximum readiness and data quality from these instruments when deployed on ASP field projects.

Alternatively in some instances the facility instruments might be second copies of the field deployable instruments. Experience would be gained in accuracy and precision of instruments by running field instrument and facility instrument side by side prior to asp field projects.

Expected outcomes of these measurements would include:

- Correlations among multiple aerosol properties.
- Patterns of new particle formation and particle growth in relation to presence and concentration of responsible atmospheric constituents. Chemical composition as a function of size and relation to pertinent gas-phase constituents.
- Changes in cloud nucleating properties of aerosols in relation to composition.

Facility would:

- Serve as a host for guest instruments, especially new methods for characterizing aerosol properties, permitting comparison with alternative measures.
- Provide calibration facility especially for particle size
- Provide opportunity for laboratory generation of aerosols under well specified conditions (or smog chamber studies), allowing these aerosols to be characterized by the pertinent subset of foregoing instruments.

Location should be subjected to a variety of aerosol types: aged continental, fairly recent urban, occasionally very clean, occasionally marine ...

ASP Ground Aerosol Observation Experiment

To investigate aerosol properties, processing/evolution, and their seasonal dependence in air masses dominated by different emission sources

Motivation- A ground measurement site at a suitable location is needed to gain understanding of aerosol properties and processing as a function of emission sources, atmospheric lifetimes, and the season. We envision that a site on Long Island (e.g., at BNL) is highly representative of a location that can sample air masses that have been exposed to different emissions depending on their trajectories: e.g., urban and industrial emissions are expected for air from the west (New Jersey and New York City) or the north (Connecticut), cleaner air from the eastern LI, and cleaner still from the south (marine). The scientific objectives will include characterizing and understanding of the following aerosol properties as a function of sources, age, and the season:

- number and size distributions, mass loading and chemical composition
- light scattering and absorption and cloud nucleation properties in relation to aerosol chemical composition
- light absorption coefficient as a function of black carbon content and aerosol processing
- Sulfate aerosol formation and processing, including neutralization by NH₃
- Dependence of SOA formation on VOC, photochemical reactivity, and aerosol acidity
- Production of POA and SOA from urban and biogenic emissions
- Effects of cloud processing
- Relationship between ground measurements and remote sensing data

Location- A Brookhaven National Laboratory site is a good compromise between having no significant local sources and the convenience of operating and maintaining the instruments. A suite of gas and aerosol instruments will be deployed to measure the following:

- Aerosol size and number distributions: pcasp and DMA
- Aerosol light scattering: 3 wavelength nephelometer
- Aerosol light absorption: 3 wavelength PSAP
- Aerosol chemical composition: Aerodyne AMS and PILS.
- Aerosol black carbon: photothermal interferometer
- Aerosol CCN properties: two DMT CCN's at different supersaturations
- Aerosol total number concentrations: two CPC's at different cut off sizes
- Concentrations of the following gases:
 - SO₂, NH₃, HNO₃, CO, O₃, NO_x, NO_y, H₂O₂
- Selected hydrocarbons, including benzene, toluene, isoprene, and monoterpenes: PTR-MS
- Meteorological parameters, such as UV irradiance, temperature, wind direction and speed, will be measured.

Collaborators- those who provide radiation measurements and remote sensing capabilities.

Carbonaceous Aerosol Evolution (CARE) Campaign

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Current state-of-the-science aerosol models do not have a reliable treatment for simulating the physical and chemical evolution of carbonaceous aerosols, which critically limits our ability to assess their direct and indirect radiative effects on climate. Carbonaceous aerosols include primary organic aerosols (POA), primary black carbon (BC) particles, and secondary organic aerosols (SOA). POA and BC are emitted from automobile exhaust, other anthropogenic activities, and biomass burning while SOA are formed via homogeneous nucleation, condensation, or heterogeneous reactions of myriad photochemical oxidation products from numerous hydrocarbon precursor gases of both anthropogenic and biogenic origins. Recent studies show that current models severely underpredict SOA formation in ambient urban atmosphere as well as in the upper troposphere ([Heald et al., 2005](#); [Johnson et al., 2006](#); [Volkamer et al., 2006](#)). To address this gap we propose the Carbonaceous Aerosol Evolution (CARE) field campaign to study the formation and aging of carbonaceous aerosols. The goal of this research is to evaluate, improve, and validate physically-based aerosol models that predict how climate-relevant properties of aerosols change as a function of time and environmental conditions (temperature, pressure, relative humidity, solar radiation, and chemical environment) so that the relevant processes can be properly treated in climate models.

Key process-level questions and knowledge gaps in carbonaceous aerosol modeling include:

- 1) Which are the most important gas-phase precursors and their oxidation products for SOA formation?
- 2) What is the rate of SOA formation in the ambient atmosphere, and how does SOA yield depend on different SOA precursors, gas-phase oxidants, pre-existing POA mass, particle acidity, temperature, and relative humidity?
- 3) What are the chemical composition, volatility spectrum, optical properties, and hygroscopicity of the carbonaceous fraction of ambient aerosols, and how do they evolve as a function atmospheric processing time and photochemical age.
- 4) What are the chemical and physical interactions between oxidation products of anthropogenic and biogenic SOA precursors, and how do these interactions affect the overall SOA formation?
- 5) What are the interactions between biogenic SOA formation processes and biomass burning aerosols?
- 6) How rapidly do SOA, POA, and BC become internally mixed with each other and with other semi- and non-volatile inorganic species, and what are the relative roles of condensation and coagulation processes?
- 7) How does the aerosol mixing state alter its optical properties, hygroscopicity, and CCN activity?

The comprehensive ground and aircraft data collected during the recent MAX-Mex campaign should provide valuable information on the formation and evolution of carbonaceous aerosol from urban, biomass burning, and biogenic emissions. However, it will be difficult to fully understand and reliably model the highly complex physical and chemical interactions between these different sources, and isolate their non-linear effects on overall SOA formation from each source. We therefore envision a series of CARE field studies of graduated complexity to complement the MAX-Mex campaign and address all of the science questions listed above and the related issues. Here, we propose the first CARE field campaign during the summer/early fall season to characterize the formation of SOA and aging of BC and POA downwind of a relatively isolated, large urban area that has well-defined and relatively uncomplicated meteorology, and which is located in a region that has little or no biogenic emissions of SOA precursors. This will allow us to focus on the evolution of anthropogenic carbonaceous aerosols without the complications resulting from biogenic SOA formation. The observational strategy will be to make comprehensive gas, aerosol, and meteorological measurements upwind, within, and downwind of the urban area with Battelle's G-1 aircraft and at strategically located ground sites. Candidate field sites for the proposed campaign include Dallas (TX), Oklahoma City (OK), and other urban areas that are relatively isolated and located within desert-like environs (e.g., Las Vegas).

The continuous fixed-location and quasi-Lagrangian datasets to be obtained will enable process modelers to perform local radiative and CCN closure studies, and to constrain, test, and validate thermodynamic and

kinetic models of SOA formation, aerosol evolution, and black carbon aging. The comprehensive datasets and knowledge gained from such detailed process-level analyses can then be integrated into 3-D models to evaluate their direct and indirect radiative effects on climate in a larger realm.

Subsequent CARE campaigns could be conducted at (a) an isolated urban area that is located within a large forested region to understand SOA formation from biogenic emissions only as well as the complex interactions between urban and biogenic emissions; (b) biomass burning/forest fire sites to understand the interactions between biogenic emissions and biomass burning carbonaceous aerosols; (c) some of the same locations as before, but in a different season.

Measurement Wish List

Key measurements include aerosol precursor gases, size-resolved particle composition, and their physical and chemical properties. Below is a preliminary list of instruments desired on the G1 aircraft and at multiple ground sites:

Aircraft (the final instrument suite will be based on need, weight, space, and power considerations)

Aerosol Mass Spectrometer

PTRMS

SMPS

Single Particle Soot Photometer

Particle In Liquid Sampler (PILS)

NO_x, NO_y, O₃, SO₂, NH₃, CO, VOC (canisters), radicals, peroxides

Nephelometer, PSAP, Photo-acoustic

Particle morphology and ¹⁴C (post-campaign analysis of field collected aerosols)

Radiation (down-welling and up-welling, spectrally resolved)

Airborne MAX-DOAS (vertical profiles of glyoxal, NO₂, HCHO)

Ground Sites

Aerosol Mass Spectrometer

SMPS

PTRMS, Chemical Ionization Mass Spectrometer (CIMS)

EC/OC Analyzer

Single Particle Soot Photometer

PILS (Water Soluble Organic Carbon, WSOC)

MAX-DOAS (glyoxal, NO₃ radical, etc., over long paths)

NO_x, NO_y, O₃, SO₂, NH₃, CO, VOC (canisters), radicals, peroxides

Nephelometer, PSAP, Cavity Ringdown, Photo-acoustic

CCN counter, HTDMA

Radiation and photolysis rates

Particle morphology and ¹⁴C (post-campaign analysis of field collected aerosols)

References

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- Johnson D. et al., Simulating regional scale secondary organic aerosol formation during the TORCH 2003 campaign in the southern UK, *Atmos. Chem. Phys.*, 6, 403-418, 2006.
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Deep Convective Clouds and Chemistry (DC3)

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Central U.S., June 2009

Science Objectives: To quantify the role of continental, midlatitude deep convection in the vertical transport, transformation, and scavenging of aerosol, aerosol precursor gases, and oxidants

Key Measurements: DC3 will provide high altitude measurements of gases, cloud condensate, interstitial aerosol, and cloud-borne aerosol detraining from deep cumulus clouds over the central U.S. This information can be combined with DOE ASP G-1 measurements of gases and aerosol entrained into the clouds at multiple levels from the boundary layer to the mid troposphere. Entrainment of gases and aerosol into the clouds would be determined by measuring the winds and concentrations in a series of circular routes around (but not in) the cloud at successively higher altitudes from the boundary layer to the middle troposphere. Entrainment at levels between the circle altitudes could be approximated by Doppler radar wind retrievals and concentrations measured between the circle levels.

Expected Findings and Implications to Climate Issues: The combination of this information can be used to determine the net scavenging efficiency (after in-cloud production) of each gas and aerosol specie. This would provide an important test of our understanding of vertical transport and wet scavenging of aerosol and aerosol precursor gases by deep convection. Although the scavenging efficiency is likely to be high (>90%), the small fraction that is detrained is important because its subsequent lifetime will be much longer than boundary layer aerosol and because both the gases and aerosol detrained from the cloud play important roles in the production of new particles as the cloud condensate evaporates in the upper troposphere. DC3 plans to apply a similar method to perform a water mass budget for the cloud.

Possible Partners: A proposal is being prepared for submission to NSF in December 2006. NOAA may also participate. The NSF HIAPER (G-V) aircraft would sample the anvil. A NASA DC8 or and the NCAR C-130 would sample the air entrained into the cloud in the upper troposphere and, if the G-1 is not available, in the lower troposphere. The ASP G-1 would sample air entrained below cloud and around the cloud in the lower troposphere. DC3 is planning to deploy a thermal tandem differential mobility analyzer and an aerosol mass spectrometer (being developed by Jose Jimenez) behind a counterflow virtual impactor (CVI) on the HIAPER. ASP could supplement this with Alex Laskin's Time-Resolved Aerosol Collector. DC3 would also measure SO₂, HNO₃, and H₂O₂. Since the focus of DC3 is on the chemistry of the anvil, it is likely to provide excellent sampling of the winds, aerosol and precursor gases throughout the anvil. The counterflow in the CVI can be turned off when flying outside the cloud so that all of the interstitial aerosol can be sampled. Current plans are to base the HIAPER and either the

NASA DC8 or the NCAR C-130 somewhere north of Oklahoma City, permitting deployments to northern Oklahoma, Northeast Colorado, or Alabama, where lightning mapping arrays and chilled radars are located. By expanding the area of the experiment, the likelihood of deep convection on any give day is increased, thus reducing idle time. Since the DOE G-1 cannot fly nearly as fast as the HIAPER or DC8, it may not be able to participate in the sampling of clouds in all three regions. DOE may prefer to base the G-1 at a different site if Oklahoma is not the study region preferred by DOE.

Ganges Valley Aerosol Experiment (GVAX)

V. Rao Kotamarthi (vrkotamarthi@anl.gov), Argonne National Laboratory

- The Asian Brown Cloud is suggested as a major contributor to the alteration of regional climate and primarily effect monsoons over India
- Recent satellite data from MISR suggests the Ganges valley is shrouded by an optically thick layer of soot/smoke much of the winter and stands out as a 'dark' in the satellite data sets he is analyzing

Objectives:

- Conduct an intensive field campaign to understand the meteorology and chemistry of the region observed from the satellites as having one of the highest optical thickness in the world.
- What is the chemical and physical composition of this airmass?
- What is its spatial extent, its source regions and the transport corridors into and out of India?
- What kind of mitigation strategies can be used in the future?
- What is the contribution of this region to the Asian Brown Cloud over the Indian Ocean?

Proposed Plan:

- A two stage experiment
- STAGE 1: A scoping experiment with mainly fixed ground station observations and portable laboratories such as the DOE ARM mobile facility.
- STAGE 2: A full scaled experiment with ground based observation stations and airborne observations from an aircraft platform, balloons and surface portable laboratories
- STAGE 1 will build preliminary data sets for more detailed modeling and building capacity on the Indian side for participating in the STAGE 2 as the primary participants and leaders.
- STAGE 1 can also be used to set the stage for scientist exchanges and identifying instrument needs on the Indian side and lay the groundwork for a sustained and more vigorous field observation programs for the future in India,
- STAGE 2 involve scientists from India, US and Europe

Facilities/Timeline

- STAGE 1 will also help identify suitable ground stations for STAGE 2 and identify power requirements.
- The STAGE 1 could be help winter 2007 and full experiment (STAGE 2) during the winter of 2008/2009.
- The Indian Government has an MOU with the DOE for collaboration on the energy sector and technology help and transfer, including environment and this could be a primary sponsor for the experiment under the aegis of this forum.
- The IMD from India and ANL/PNNL from US for ground meteorological observations.
- The MRWF (Noida, India) and IITM (Pune) as the primary Indian modeling support and ASP program in the US for the same role. University scientists from India, ASP and NCAR scientists for Chemical and aerosol measurements.

Indirect and Semi-Direct Aerosol Campaign (ISDAC)

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North Slope of Alaska, April 2008

Science Objectives:

1. Assess our understanding of the activation of arctic aerosol as cloud condensation nuclei and as ice nuclei.
2. Assess our understanding of the influence of arctic aerosol on droplet number, crystal number, and glaciation of arctic clouds.
3. Assess our understanding of the mechanisms for scavenging of arctic aerosol.

Key Measurements: Counterflow Virtual Impactor to isolate activated aerosol from interstitial aerosol, Cloud Condensation Nuclei (CCN) spectrum, Ice Nuclei (IN) spectrum, updraft velocity, single-particle composition, and size distributions of aerosol number, aerosol composition, aerosol hygroscopicity, droplet number, drizzle, and crystal number.

Expected Findings and Implications to Climate Issues: We expect to find that we have much to learn about cloud-aerosol interactions in the arctic, where aerosol concentrations are high during winter and early spring, aerosol indirect effects are known to operate for both shortwave and longwave radiation, and where climate sensitivity is extremely high. The highly aged nature of arctic aerosol during winter will put our understanding of internally-mixed aerosol activation to the test. The high frequency of mixed-phase clouds will stress our understanding of ice nucleation and glaciation of clouds. The apparent scavenging of arctic aerosol during spring is a mystery that has profound implications for shortwave indirect effects (which would be much stronger during summer if the aerosol were to survive). A CVI has previously been coupled with an aerosol mass spectrometer (Cziczo et al., 2004) and single particle analysis (Twohy and Poellot, 2005) to determine the microphysical and chemical properties of IN in the tropics. This experiment would provide the first direct measurements of the microphysical and chemical properties of activated CCN and IN and of scavenged aerosol in the arctic. The experiment will provide data for CCN, IN, and droplet number closure experiments and for evaluating cloud-aerosol interactions in cloud model and cloud-aerosol model simulations.

Possible Partners: A proposal for much of this experiment has been submitted to the ARM program, with a decision due by September. ARM would provide an aircraft that would provide high-quality cloud measurements. ASP would provide the G-1 to measure the properties of the aerosol below cloud and the residual particles evaporated from droplets and ice crystals. The ARM aircraft could be the G-1 or a separate aircraft.

References

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Twohy, C. H., and M. R. Poellot: Chemical characteristics of ice residual nuclei in anvil cirrus clouds: evidence for homogeneous and heterogeneous ice formation. *Atmos. Chem. Phys. Discuss.*, 5, 3723–3745, 2005

Megacity Aerosol Experiments

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Background

It is predicted that by 2025 two thirds of the Earth's population will live in urban areas, with many living in "Megacities" which have more than 10 million people. Ninety percent of the growth in urban population will occur in developing countries. For many of these areas there is virtually no information on the climate altering pollutants that they export to the global environment. Given differences in land type, economic activity, fuels, and transportation, Megacities in developing countries are expected to emit aerosols that differ significantly in amount and properties from that observed in developed countries. A large aggregate impact from Megacities can be inferred from measurement campaigns such as INDOEX, in which the radiative impact from aerosols generated on the Indian sub-continent was quantified and found to be quite large.

By and large the air chemistry community has chosen to study the impacts from developing countries by making measurements hundreds to thousands of km from pollution sources (i.e. INDOEX, TRACE-P, ACE-Asia). There are a few exceptions including the recently completed Mexico City MILAGRO campaign and a much smaller scale surface survey to be conducted in Mumbai, India and Lagos, Nigeria by a NSF group led by Dan Reimer, University of Miami. There is an on-going planning process, under the IGAC umbrella, to extend MIRAGE (NSF component of MILAGRO) to other Megacities. It is unlikely that this will occur before 2010.

The United States may be excellent laboratory for studying aerosol processes, but in order to form a picture of global impacts relevant to climate change increased attention must be paid to locations that are projected to be the major contributors to the planets anthropogenic aerosol burden. It can't all be done with satellites, models, and stand-off downwind observations. It is inevitable that the need for information about aerosol source regions will drive the climate and air chemistry communities to measurement campaigns in Megacities in developing countries.

There are many logistic difficulties to making these measurements. Designing a suitable campaign takes time. It is appropriate that the DOE Atmospheric Science Program begin making plans. What follows are some thoughts on what is important and how to proceed.

Criteria for Selecting a Location

- _ Areas thought to be characteristic of important aerosol emitting regions.
- _ Emissions and air chemistry are poorly characterized.
- _ Safe passage, logistical support, and something approaching 24/7 electricity.
- _ Permission to enter airspace.
- _ Emerging technologies, such as ethanol use in Sao Paulo and Rio de Janeiro, Brazil

- _ Areas that have been studied downwind, i.e. India, China
- _ Collaborations

Objectives

- _ Characterize aerosol composition, size distribution, optical properties, and CCN properties.
- _ Determine modifications to clouds.
- _ Link aerosols to primary sources and precursor emissions and then to economic and life-style activities.
- _ Determine if aerosol chemical and physical transformations fit into the framework contained in models - derived largely from observations in the Industrialized world.
- _ Evaluate representativeness of measurements from satellite observations. Provide ground truth for satellite retrievals.
- _ Determine relations between near-source measurements and those obtained hundreds or thousands of km distant in stand-off campaigns
- _ Quantify radiative impacts of aerosols from direct and indirect effects. Quantify anthropogenic component.

Possible Types of Campaigns (in order of increasing complexity):

Survey:

Deploy a surface site in select Megacities for 2 to 4 weeks. Possibly a mixture of fixed base sampling and a roving deployment, such as used by NSF. Limited air quality information should not be confused with zero information. The challenge is to come up with a do-able instrument package that is a qualitative advance over what already has been done. Although logistics are always a major concern, it may be possible to do survey measurements without relying on the participation of scientists from the host country. Such measurements would focus on characterization and could be a preliminary phase to a more complex experiment.

Ground Site:

Surface measurements of a broad range of aerosol, trace gas, and possibly radiative properties. As the complexity increases, participation of scientists from the host country becomes increasingly important. Measurements would address characterization and process objectives.

G-1 field program:

The G-1 provides additional instrumentation and a wider footprint for measurements. Downwind measurements would explicitly show the time evolution of aerosols and their contribution to the global atmosphere. Planning process should weigh the options of a field campaign consisting mainly of ASP investigators versus a larger multi-agency effort.

MILAGRO type Campaign

Programs of this scope require a lead time of order 5 years. There has to be coordination among multiple agencies as well as negotiations with foreign governments. Participation of scientists from the host country is essential. ASP activities would include surface and aircraft sampling.

How to Proceed

- _ Identify ASP objectives and venues where these objectives can be addressed.
- _ Continue discussions with global climate modelers to identify their needs.
- _ Bring satellite remote sensing community into discussions.
- _ Assess resource requirements.
- _ Find partners in other agencies (and other countries) with overlapping objectives.
- _ Participate in MIRAGE and other international planning exercises.
- _ Form collaborations with scientists in host countries.

Rapid deployment capability for characterization of unique events

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At the BERAC committee meeting it was asked whether ASP is ready to take advantage of unique opportunities to sample and follow and characterize unique events such as volcanic eruptions.

Such capability would require availability of suitable aircraft, equipment, and personnel. Such capability would have to draw on existing capabilities. For example instruments and personnel might be rapidly deployed if ASP had an ongoing aerosol characterization facility.

Examples of possible events:

Upper troposphere - lower stratosphere: volcanic

Lower troposphere: e.g., major fires, volcanos, natural or human induced events e.g., WTC, Kuwait oil fires. Radiological, e.g., Chernobyl.

Required capabilities

In-situ measurements: Aircraft. Usual complement of chemical and microphysical instruments.

Remote sensing: Aircraft Lidar, Satellite Passive and Active. Need a satellite person or liaison

Meteorological forecasting

Resource requirements

Would require additional copies of instruments maintained in ready to go status; calibrated; ideally aircraft should be located within one day of aerosol facility.

Personnel resources. Share with aerosol facility.

The Biospheric Aerosol Production Experiment - BAP

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The subject field campaign will investigate the production of secondary organic aerosol (SOA) through chemical processing of biogenic nonmethane organic compound (BNMOC) emissions from temperate forests. On a global scale, approximately 90% of the NMOOC emissions originate from biogenic sources. About 50 % and 10% of the BNMOC emissions are composed of isoprene (a hemiterpene) and monoterpenes, respectively. Terpenes are very reactive, much more so than the NMOOCs emitted from anthropogenic sources. Monoterpenes have been identified as major contributors to SOA production in forested areas and isoprene has been postulated to be an important SOA precursor emitted by deciduous forests. The global production of SOA through chemical processing of BNMOCs is comparable in magnitude to the amount of sulfate aerosol produced by natural and anthropogenic sources combined and ten times the global production of carbonaceous soot. Biogenically derived SOA could affect climate by scattering and absorbing solar radiation and by serving as cloud condensation nuclei. The physical, optical, and chemical properties of biogenic SOA at, near, and downwind of source regions are highly uncertain and thus require systematic analysis.

The scientific objective of the BAP field campaign is to rigorously evaluate the production of SOA in a globally significant forested ecosystem. There is sufficient evidence from chamber studies to suggest that SOA formation can involve homogeneous and heterogeneous nucleation of BNMOC reaction products and particle growth through condensation of various gas-phase organic compounds. However, the very few field studies of SOA production conducted in forested areas to date have been small in scope and have been unable to confirm the proposed physicochemical mechanisms or to evaluate SOA production rates and yields in the atmospheric boundary layer (ABL). We propose an intensive, month-long field campaign in which diurnal measurements of chemistry and meteorology are made from within and above a temperate forest using surface sites, instrumented towers, tether sondes, and aircraft. Possible field sites include forests in NE Texas, the Ozarks, the Sierra Nevada, and Nova Scotia.

The ultimate goal of the BAP field campaign is to better understand the formation of biogenic SOA and their role in direct and indirect radiative forcing of climate. Spatial and temporal variations in the composition and concentrations of carbonaceous aerosol have confounded efforts to evaluate their effects on radiative forcing. Thus, the approach of the BAP field campaign will be to make high-frequency measurements of SOA precursors, oxidants, and gaseous- and aerosol-phase reaction products at and above the surface. A complete process scale understanding of the production of SOA from biospheric emissions is crucial for development of the next generation of coupled climate models, which are expected to include dynamic vegetation models. The robust data set composed of vertical-profile and high-temporal-resolution measurements of the carbonaceous aerosol concentrations and composition in the ABL will be used by ASP scientists, (1) to better understand the chemical pathways of SOA production, (2) to estimate biogenic SOA production rates and yields in the atmosphere, (3) to evaluate the effects of biogenic SOA composition on the light-scattering and light-absorbing properties of carbonaceous aerosol (i.e., the direct radiative forcing of climate), and (4) to evaluate the effects of biogenic SOA composition on surface tension, CCN production rates, and activation of CCN to form clouds (i.e., the indirect radiative forcing of climate).

PROPOSED G1 EXPERIMENT

Time evolution of the optical and chemical properties of smoke from brush and pine forest fires in the relatively dry western US.

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Background: Significant changes of the optical coefficients, single scattering albedo, and chemical composition of smoke from biomass burning in Amazonia and southern Africa have been observed (1, 2) on time scales ranging from minutes, to hours, to days. For example, aerosol single scattering albedo at 550 nm was observed to change from 0.84 to 0.90 in 5 hours for smoke in southern Africa, and the mechanism of change was attributed to gas to particle conversion that added scattering material to particles (1).

Recent laboratory measurements supported in part by DOE-ASP on smoke optical properties revealed strong dependence of the aerosol single scattering albedo and Angstrom coefficients for absorption and scattering on fuel type and on whether flaming or smoldering combustion conditions are present. Smoke from combustion of sage brush or chamise, two common shrubs in the west, showed single scattering albedos as low as 0.3 at 532 nm during the flaming stage. In addition, chamise smoke single scattering albedo doubled when the RH was changed from dry conditions below 30% to 85%, and particle morphology collapse from 30 nm monomers chain aggregated with a fractal dimension of around 2 to more compact clusters with fractal dimension closer to 3. Absorption decreased upon collapse by 30%, and scattering was increased. Chamise smoke had a substantial inorganic composition that likely determined water uptake. It is likely that the process of adsorption added water molecules to the particles at RH below 55% in some cases, and that the adsorbed water vapor supplied enough tension to initiate collapse. It appears that processing of smoke at relatively modest RH values can substantially affect aerosol optics. Figures 1 through 3 summarize the laboratory findings. Figures 1 and 2 show the strong variation of single scattering albedo with fuel type at 405 nm and 532 nm (horizontal axis) and the substantial increase of Angstrom coefficient for absorption calculated between these wavelengths and 870 nm as the single scattering albedo increases and light absorption by organic material is observed. Figure 3 shows the reduction of aerosol absorption with RH likely due to particle collapse and shielding of some of the black carbon from light absorption as the electromagnetic skin depth is decreased.

The laboratory results can be roughly summarized as follows. Smoke from bushes and shrubs can produce very low single scattering albedos. Smoke from grasses has high single scattering albedo and substantial fraction of absorption due to organic species, in addition to elemental carbon. The inorganic content of smoke strongly determines whether particle collapse will occur at RH well below 100%.

Climate impacts of smoke need to be modeled with proper treatment of aerosol optics and chemistry. Remote sensing retrievals of aerosol optical properties that assume simple wavelength dependencies for light absorption can be substantially in error if the laboratory observations are also observed in nature. Laboratory combustion conditions perhaps are less

oxygen limited than large wildland fires, so perhaps more smoldering combustion is observed in nature than in the lab.

FIELD PROGRAM: Fly the G1 out of the Stead airport near Reno, and take advantage of the Desert Research Institute and the University of Nevada Reno as command and control centers for air and ground based activities. DRI has a strong program in wild land fire, including fire weather prediction, so take advantage of forecasts from this program for smoky conditions, field campaign design and timing, and analysis. Reno Nevada is in an area where wildland fires are very common in summer months during the dry season, and a variety of fuel stocks burn, from grasses, to sage shrubs, to pine forests. Prescribed fires are also used to clear under brush in an attempt to mitigate natural fire intensity should one occur in the future.

Smoke in the western US may be impacted by only modest RH conditioning, RH levels below 85%, though laboratory measurements have shown substantial impacts on smoke optics even at these levels. Smoke aerosol seems to generally become ‘whiter’ (increased single scattering albedo) with age due to particle aggregation, morphology change with RH processing, and gas to particle conversion. Which effect is most important in relatively dry areas, and what are the relevant time scales for transformation of aerosol optics and chemistry? What are the differences observed with different fuels, especially grasses, shrubs, and pine forests? In summary, what processes govern the variation and evolution of aerosol single scattering albedo downwind of biomass burning events in relatively dry areas with a wide variety of fuel stocks?

Key measurements include gas phase chemistry to measure combustion efficiency near sources and transformations. Instruments should include, but perhaps not be limited to the following:

1. Mass spectrometer for real time particle chemistry.
2. Photoacoustic absorption with nephelometer built in for fast response aerosol optics.
3. Multiwavelength photoacoustic measurements (405 nm and 870 nm) to evaluate spectral tendencies, especially absorption in the UV by OC species.
4. Cavity ringdown aerosol extinction and TSI neph scattering for closure measurements of aerosol absorption when single scattering albedos are sufficiently small for this technique to be accurate.
5. Radiometric package including sun photometers to get column properties and in situ extinction by vertical differentiation.
6. Good RH, wind, and CN sensors to use in tracking smoke plumes.

Flight strategies will follow plumes from very near their sources down wind, both in time and space. Wild land fires quickly move through their flaming stages (hours) and may smolder for days, depending on fuel, environmental conditions, and fire management strategy. Vertical profiles will also be used to quantify smoke extent and RH conditioning of smoke.

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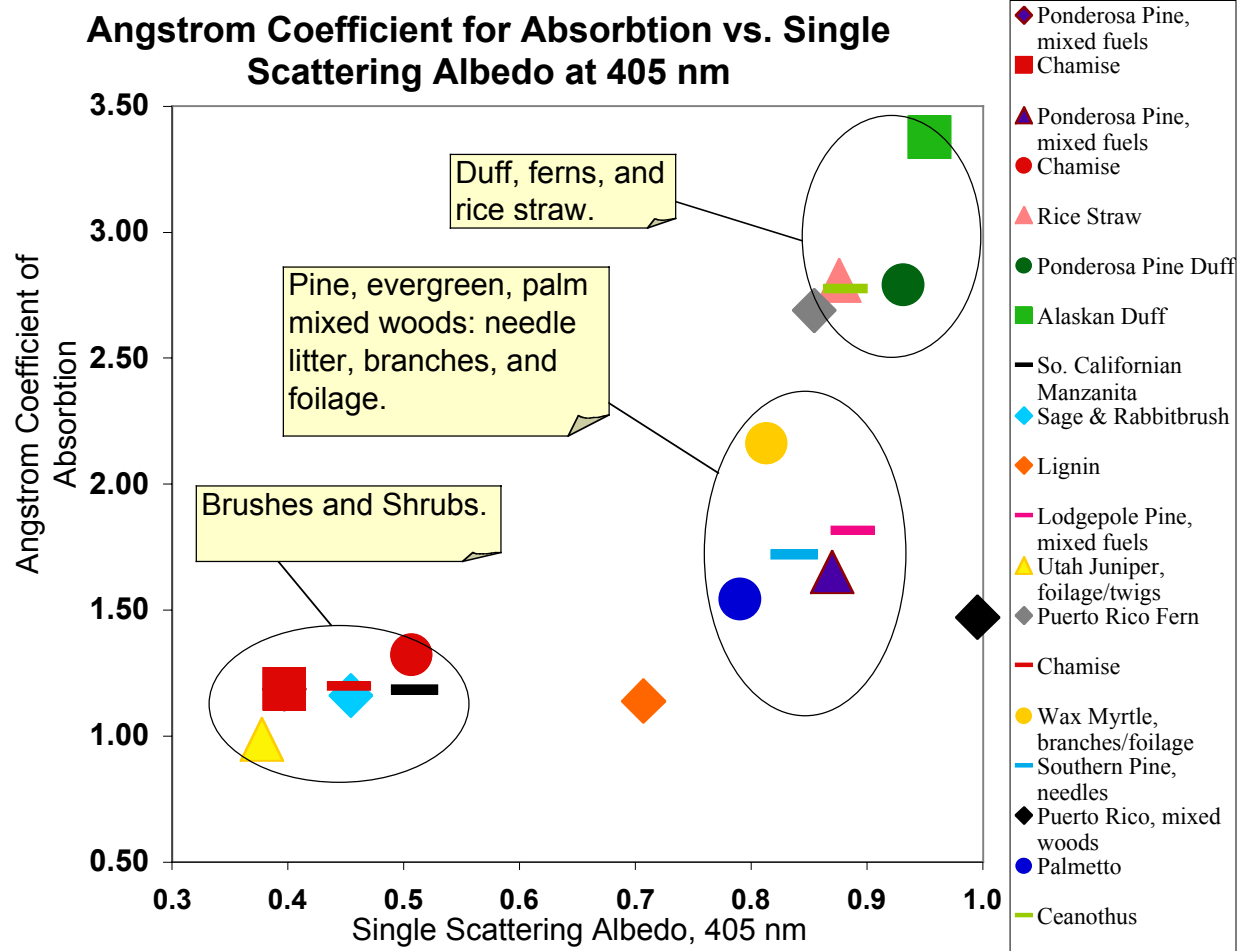


Figure 1. Variation of the Angstrom coefficient for absorption with single scattering albedo at 405 nm as measured by the 2 wavelength photoacoustic instrument, 405 nm and 870 nm. The 870 nm wavelength absorption measurement is likely due to the elemental carbon fraction while at 405 nm substantial absorption by organic material is present. Note the wide range of single scattering albedo.

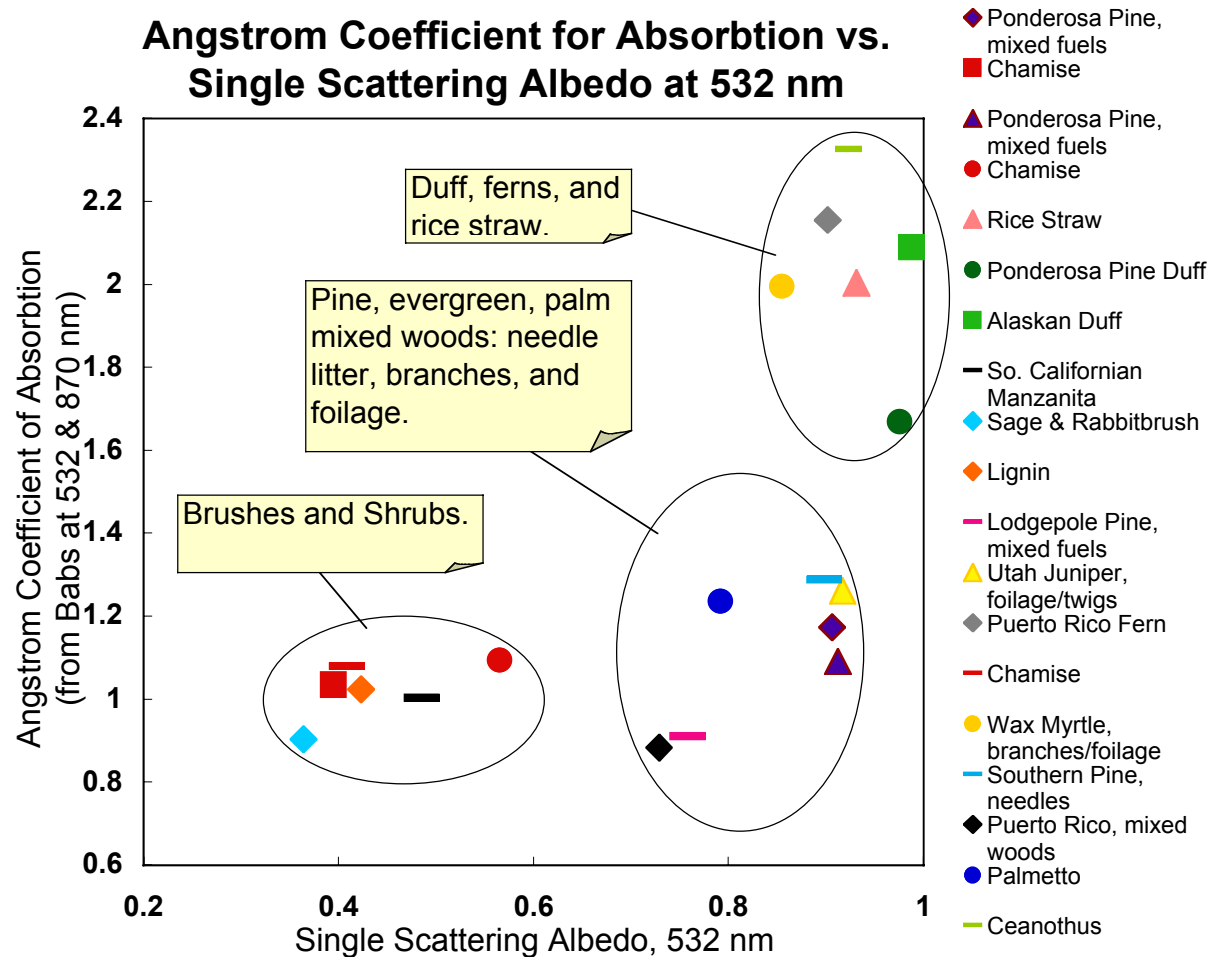


Figure 2. Same as figure 1, however the wavelength here is 532 nm instead of 405 nm.

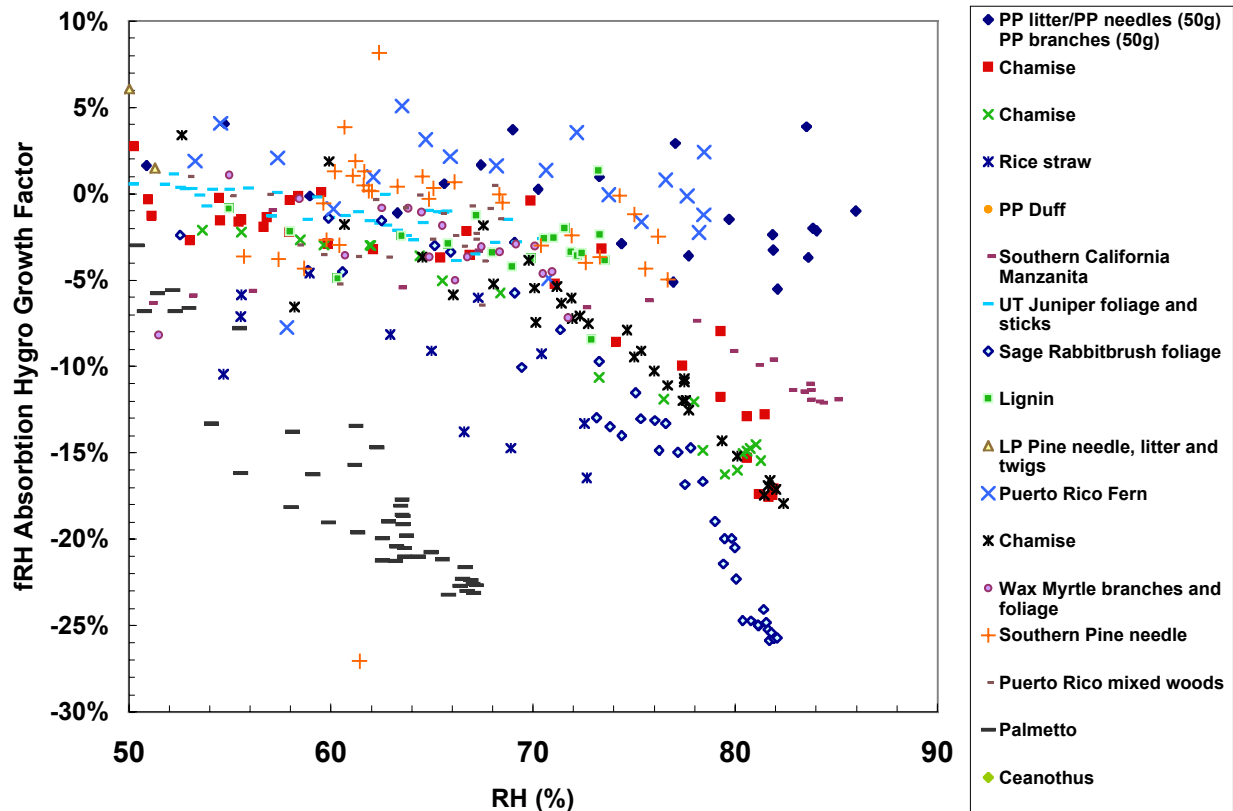


Figure 3. Variation of the hygroscopic growth factor for aerosol light absorption with relatively humidity. Negative values correspond to reductions of absorption with RH increase, likely due to particle collapse with increasing RH. The amount of reduction depends strongly on inorganic content. For example, palmetto smoke had the highest ratio of inorganic to organic mass (strong change of absorption with RH), and pine had one of the lowest (no change of absorption with RH).

Winter/Summer Aerosol Composition and Evolution Experiment

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It is proposed that major aircraft based clear-air experiments be conducted in the winter of 2008 with a follow on study in either the summer of 2008, or more likely the summer of 2009. It is proposed that these two experiments be conducted in the same region of the US so that advantage can be taken of the seasonal differences in aerosol precursor source strengths, and processing conditions.

Motivation- The formation and processing of atmospheric aerosols represent a major uncertainty in the modeling of aerosol distributions that is needed to assess the radiative forcing of ambient aerosol particles. A particular need is to develop an understanding of the processes that lead to the formation and evolution of secondary organic aerosol (SOA) although it is also important to develop further understanding of the details of the processes forming aerosol sulfate and nitrate under different sets of conditions.

Most climate-related studies of aerosol formation and growth processes have been conducted during the summer when photochemistry is active and biogenic sources of organic aerosol precursors are at their maximum. With the exception of studies focused on health effects, there have been few detailed studies that have been conducted in the winter. The lack of knowledge regarding aerosol composition, formation and growth in the winter is in itself sufficient reason to conduct a winter experiment. But, it is our view that comparison of summer/winter data may give valuable information with regard to the relative importance of various precursors, and much needed insight into the processes that lead to the accumulation of organic aerosol mass, particularly if such studies were carried out at the same location.

Summer-winter differences likely to be important include-

1. Rate of formation of aerosol mass- Rate of accumulation of aerosol mass by photochemical processes will be reduced in winter relative to summer because of reduced solar intensity and generally lower absolute humidities that will reduce OH concentrations. Reduced OH and will suppress peroxide formation in winter reducing the importance of aqueous-phase sulfate production. Distribution of semi-volatile substances to aerosols should be enhanced because of reduced temperatures in winter relative to summer.
2. Aerosol composition-Difference in emissions (e.g., reduced biogenics) should result in changes in aerosol composition. There is some evidence that nitrate is a more important aerosol component in winter relative to summer.

Differences in composition may lead to differences in optical properties, CCN activity, etc.

3. Residence time of aerosols should be enhanced in winter relative to summer because of reduced vertical mixing. In combination with the generally higher winter wind velocities, aerosols from a given set of sources should be more widely dispersed.

Scientific Questions-

Scientific issues to be addressed include:

- POA source strengths of representative anthropogenic and biogenic sources
- Relationship between POA and black carbon in urban emissions
- Processing of POA in relation to photochemistry
- Formation of SOA in urban and power plant plumes in relation to photochemistry
- Formation of SOA from biogenic emissions with and without urban and power plant influences
- Hygroscopicity, light scattering and absorption coefficients, and CCN properties of aerosols as a function of sources and atmospheric age, and season.
- Growth mechanism for SOA (i.e., condensation or Pankow-Seinfeld volume growth) as a function of processing conditions including summer/winter differences.
- Evolution of aerosol specific absorption as the aerosol ages.
- Winter/summer differences in aerosol precursors particularly biogenic precursors and their influence on aerosol composition.
- Whether current aerosol models can capture the seasonal differences in aerosol composition, particularly SOA.

Experiment Strategy

The experiment should be located in a region where the aircraft can sample a significant distinguishable urban source. Presence of isolated SO₂ point sources such as power plants will be essential for both studies of sulfate formation as well as the formation of organic aerosol (acid catalysis). The study should be located where there is a strong seasonal difference in the emissions of biogenic aerosol precursors such as isoprene and pinenes.

The plan is to use the DOE G-1 aircraft as our primary sampling platform. Our flight strategy would be similar to strategies employed in similar studies in the past. Flights will be made upwind and downwind of sources of various types using a pseudo-Lagrangian strategy to track the evolution of aerosol properties as the plume ages and transformations occur. Flight plans will be developed to sample single sources and mixtures of sources of aerosols and aerosol precursors. Sources of interest include urban sources, powerplants, regions of high biogenic emissions, etc. The experiment would be

greatly enhanced by the addition of a surface site with a comprehensive array of instruments for measuring trace gases and a range of aerosol properties. Experience has shown that such measurements provide a valuable context for interpretation of aircraft data which by their nature are very limited in time.

Potential Venues Collaborative effort with other organizations to supplement data from ASP sources is highly desirable. Possibilities include a study based in the NE US similar in scope to ICARTT 2004. If we based near Boston it would be possible to sample the Boston urban plume as it is transported out over the North Atlantic. This is a real advantage because the complexities induced by the admixture of fresh aerosols and aerosol precursors as the plume ages, is minimized. A wintertime deployment in the NE would coincide with the winter experiment that is planned by NOAA for the winter of 2008.

Another possibility is to base in Nashville, TN. The latter is interesting because of our previous experience in the area, the dominance of biogenic organics in the summer, and the presence of large isolated powerplants in the area that will provide a natural laboratory for the study of winter vs summer sulfate formation. A summer/winter comparison based in Nashville would also be particularly useful for understanding the importance of biogenics as a source of organic aerosols. This region, because of its forest/vegetation coverage, has been extensively studied during the Southern Oxidant Study in the 90's focusing on identifying the role biogenic hydrocarbons play in photooxidant formation. DOE through its ACP program was a major participant, contributing many important scientific findings concerning ozone production in urban and power plant plumes. Several important considerations that make this location desirable are:

- Nashville is a fairly isolated urban area whose emissions as well as regional background can be well identified, tracked, and characterized.
- This region is part of the forested southeast of the US where biogenic hydrocarbon emissions, both isoprene and monoterpenes, are highly important
- The terrain of this region is relatively flat, simplifying the mixing characteristic the plumes
- Nashville and its vicinity have at least four major power plants, one located at the edge of the metropolitan area, the other three are located at ~100 km to the north and west.
- A wide range of conditions concerning emission sources and photochemical productivities can result depending on wind directions

Other possible locales for an experiment include an urban area in the Midwest such as Columbus, OH which has a modest population (800K, 1800K metropolitan); Chicago (>5000K depending on what's included); Minneapolis-St. Paul (3,000K) is an interesting possibility because of it's isolation from other urban areas.

Potential Collaborations. NOAA is planning a winter experiment in the NE in the winter of 2008 with a base in Portsmouth, NH. Could locate G-1 there and take advantage of resources that NOAA and other organizations will bring to table. Scientists from the Canadian Meteorological Service have also expressed an interest in working with us at just about any location. Other collaborations would have to be developed.